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Multi-objective Bayesian Optimization of Super hydrophobic Coatings on Asphalt Concrete Surfaces

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Abstract

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Keywords

Sustainable airfield pavement, Superhydrophobic coating, Polytetrafluoroethylene, Surrogate modeling, Multi-objective Bayesian optimization

Disciplines

Civil Engineering | Computational Engineering

Comments

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ACCEPTED MANUSCRIPT

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Abstract

Conventional snow removal strategies add direct and indirect expenses to the economy through profit lost due to passenger delays costs, pavement durability issues, contaminating the water runoff, and so on. The use of superhydrophobic (super-water-repellent) coating methods is an alternative to conventional snow and ice removal practices for alleviating snow removal operations issues. As an integrated experimental and analytical study, this work focused on optimizing superhydrophobicity and skid resistance of hydrophobic coatings on asphalt concrete surfaces. A layer-by-layer (LBL) method was utilized for spray depositing polytetrafluoroethylene (PTFE) on an asphalt concrete at different spray times and variable dosages of PTFE. Water contact angle and coefficient of friction at the microtexture level were measured to evaluate superhydrophobicity and skid resistance of the coated asphalt concrete. The optimum dosage and spay time that maximized hydrophobicity and skid resistance of flexible pavement while minimizing cost were estimated using a multi-objective Bayesian optimization (BO) method that replaced the more costly experimental procedure of pavement testing with a cheap-to-evaluate surrogate model constructed based on kriging. In this method, the surrogate model is iteratively updated with new experimental data measured at proper input settings. The result of proposed optimization method showed that the super water repellency and coefficient of friction were not uniformly increased for all the specimens by increasing spray time and dosage. In addition, use of the proposed multi-objective BO method resulted in hydrophobicity and skid resistance being maximally augmented by approximately 23% PTFE dosage at a spray time of 5.5 s.

Keywords: Sustainable airfield pavement; Superhydrophobic coating; Polytetrafluoroethylene; Surrogate modeling; Multi-objective Bayesian optimization.

Introduction

Maintenance of roadways and paved areas of airports during winter has been a difficult task for airport agencies and highway authorities, particularly since conventional snow-removal methods like deploying snow removal vehicles and spraying de-icing agents on the surfaces are labor intensive and usually require temporary closure of airport operations (Baskas 2011; Nahvi et al. 2018; Shen et al. 2017). Needless to say, such de-icing chemicals can also cause damage to both rigid and flexible pavements (Anand et al. 2017; Merkert and Mangia 2012).

To overcome winter maintenance-related problems, emerging technologies such as super-hydrophobic coatings have received recent attention (Arabzadeh et al. 2017; Von Baeyer 2000) for use in preventing or limiting ice and snow formation. "A surface is superhydrophobic when the contact angle of droplets deposited on it is equal to or bigger than 150°"(Feng and Jiang 2006; Zhang et al. 2008). Also, it is worth mentioning that hysteresis contact angle (CAH) is another means that can be used to characterize the degree of water repellency of a surface (He et al. 2004), the smaller the CAH the higher the degree of water-repellency. In such a situation, droplets hitting the surface can easily roll off and not tend to wet the surface, and combining this effect with surface roughness and low surface energy results in superhydrophobicity (Onda et al. 1996; Zhang et al. 2008). While the main goal of application of these materials on paved surfaces is to prevent formation of ice and snow to facilitate removal operations, there is no benefit in applying materials on a pavement surface passengers are endangered by a decreasing skid resistance of paved areas under dry conditions. Therefore, after application of super-water-repellent materials, skid resistance of a pavement surface must be controlled.

According to the literature, superhydrophobic surfaces can be fabricated using different methods such as: layer-by-layer (LBL), wax solidification, lithography, polymer conformation, vapor deposit, sublimation, plasma treatment, etc. (Zhang et al. 2008). Among the mentioned methods, LBL is the most suitable, because of its practicality for field implementation which has been well documented in other studies (Arabzadeh et al. 2016a, 2017; Nascimento et al. 2012). were the first to produce water-repellent asphalt concrete and they had a huge contribution in this field. It is possible to use spray deposition techniques for producing superhydrophobic coatings on asphalt concrete surfaces (Segundo et al. 2018; Zakerzadeh et al. 2018) by depositing the low surface energy materials in a single layer. Also, there are studies using LBL method to create an asphalt concrete surface coating with polytetrafluoroethylene (PTFE), and those studies used microtribometer-based coefficient of friction (CF) measuring methods to characterize the skid resistance of super-hydrophobic coated surfaces. According to the literature, spray time and dosage are significant factors affecting superhydrophobicity and CF, and if these factors are properly chosen in PTFE usage, coated asphalt can provide high skid resistance at low speeds (Arabzadeh et al. 2016b, 2017). Adel M.A. Mohamed et al (Mohamed et al. 2014) employed the Taguchi method to rank several factors that may affect the superhydrophobic properties in order to formulate the optimum conditions. Their results demonstrated that ZnO content has the highest contribution on water contact angle. Additionally, by considering the input factors independent from each other, they found the optimum design of coating surface with the water contact angle of 159° and sliding angle of 2°.

Interactive effect of spray time and dosage on asphalt concrete with respect to hydrophobicity and friction coefficient have not been well-studied, so it seems worthwhile to model the behavior of super-hydrophobic coatings on asphalt concrete surfaces while varying these two most important operating parameters (i.e., spray time and dosage). This modeling requires an expensive experimental procedure to evaluate the design objectives (i.e. superhydrophobicity, skid resistance, and cost) as functions of operating parameters, and since outcomes using experimental methods cannot be explicitly measured, the traditional optimization methods may not be employable. In the past decades, surrogate-based approaches have attracted intensive attention. These approaches can approximate and replace the expensive experimental or computational procedures with simple analytical models (Amani et al. 2017; Daghighi et al. 2017; Jansson et al. 2003; Mockus 1994; Rasmussen 2004; Simpson et al. 2001; Sun and Betti 2015). The simple model is often called surrogate; and the procedure of constructing a surrogate is called design of experiment (DoE). After constructing the cheap-to-build surrogate, optimization methods can then be applied to search for the optimum, referred as surrogate-based design optimization.

One of the powerful tools for the optimization of the design choices is Bayesian optimization (BO) which is gaining great popularity in the last decade (Shahriari et al. 2016). Fundamentally, BO is a sequential surrogate-based approach that first builds a prior belief over the possible objective functions and then sequentially refine the surrogate by observing new experimental outcomes. The new experimental observations are intelligently managed by defining an acquisition function that maximizes the probability of gaining information during the sequential updating procedure (Jones et al. 1998). To this aim, during the last few decades several acquisition functions have been proposed, e.g., Thompson sampling (TS), probability of improvement, expected improvement (EI), upper confidence bounds, and entropy search (ES) (Shahriari et al. 2016). These acquisition functions trade off exploration and exploitation criteria; in exploration criterion, the uncertainty in the surrogate model is quantified and in the exploitation criterion, the magnitude of the model prediction is quantified. BO algorithms then select the next query point by maximizing such acquisition functions. Improvement-based acquisition functions favor points that are likely to improve upon the prior believe over the objective function. The reader is referred to the (Shahriari et al. 2016) which summarized the existing well-known acquisition functions for BO.

Unfortunately, the traditional acquisition functions such as EI and PI are considered the multiple output as statistically independent. With respect to the multivariate surrogate construction, this study implements a new acquisition function based on the definitions of the aforementioned exploration and exploitation criteria.

Another key element in BO process, is the selection of proper surrogate construction technique. A large number of surrogate construction techniques have been proposed in the literature and are summarized in what follows. The dimension reduction (DR) method has been developed based on an additive decomposition that simplifies a single high-dimensional objective function to multiple one-dimensional functions (Rahman and Xu 2004). Stochastic spectral methods such as the polynomial chaos expansion (PCE) method decomposes the objective function into a set of orthogonal stochastic polynomials composed by the random inputs (Choi et al. 2004).This decomposition results in a stochastic surrogate that provides a compact and appropriate approximation of the objective function. Although, these techniques are able to handle the approximation of objective functions in high-dimensional spaces, they lack the capability of capturing the high interdependency among the design variables (Rasmussen 2004). On the other hand, Kriging, or Gaussian process (GP), is another technique of surrogate construction for which the approximations are modeled by a GP derived by proper covariance (Rasmussen 2004) The authors have previously demosntrated that Kriging has strong benefits when it comes to processing data with a small number of sample points, in low-dimensional spaces, and/or when the objective function shows a highly nonlinear behavior (Sadoughi et al. 2017). A comprehensive and detailed review of different surrogate construction techniques has been provided in (Chen 2016). Since our target objective function is expected to be highly nonlinear and is defined in low-dimensional space, Kriging is selected in this study as the surrogate modeling technique.

Similar to our case study, in practice, a given optimization problem, can involve more than one objective function. The literature on BO and surrogate construction, however, often reduces these multiple objective functions to a single function (either ignoring all the other objects or combining all objects through a weighting function) (Doh & Lee, 2018; Parussini, Venturi, Perdikaris, & Karniadakis, 2017; Qin & Faber, 2012; Rana, Li, Gupta, Nguyen, & Venkatesh, 2017; Shin & Jun, 2015). For instance, Kleijnen et al (Kleijnen and Mehdad 2014) fit univariate Kriging models for each of the two objective functions (namely, cost and service) of a call-center simulation. Constructing separate and independent univariate GPs over the objective functions may risk the loss of information related to the correlation among the objective functions. This loss can be particularly more significant, when the objective functions are somehow related to each other and thus a strong interdependency among the response surfaces are expected. For instance, in this study, we expect that the superhydrophobicity and skid resistance are highly interdependent. In order to capture the independency among the objective functions during the process of surrogate construction, this study investigates the usage of multivariate GP which constructs one joint surrogate over all the objective functions. For each candidate point in design space, the multivariate GP provides the mean value of objective function prediction and also the covariance matrix, quantifying the uncertainty of prediction and the correlation among the objective functions. Unfortunately, the traditional acquisition functions such as EI and PI are considered the multiple output as statistically independent. With respect to the multivariate surrogate construction, this study implements a new acquisition function based on the definitions of the aforementioned exploration and exploitation criteria. For the exploration criterion, a cross validation technique is adopted to quantify the nonlinearity of the objective function and prediction uncertainty over the design (or input) space, and for the exploitation criterion, the L2 norm of the mean prediction by multivariate GP is considered to measure the magnitude of the objective functions over the design space.

In summary, this study's surrogate model is based on multivariate GP and captures the interactive effects of spray time and PTFE dosage on superhydrophobicity and skid resistance. Since an important objective of such experiments is to maximize the superhydrophobicity and skid resistance in a cost-effective manner, the objective of this study is to construct a joint surrogate model that assists BO in minimizing the spray time and PTFE dosage while also maximizing the superhydrophobicity and friction coefficient.

Methodology

Figure 1 describes the general process followed in this study. The super-hydrophobicity and skid resistance of coated asphalt concrete surfaces were measured as implicit functions of PTFE dosage and spray time (input or design variables). While this study aims to maximize the hydrophobicity and skid resistance of coated asphalt concrete, cost effectiveness is another important factor in providing affordable coating surfaces for agencies, so a cost factor was introduced as following:

$$C = \frac{1}{St \times Pd} \quad (1)$$

Where C is a cost factor, S_t is a spray time and P_d as PTFE dosage-percentage. To maximize the hydrophobicity and skid resistance of coated asphalt concrete, this study proposed a multi-objective BO approach to finding the optimum and cost-effective spray time and PTFE dosage. The experimental procedure and the BO model are explained in the following sections.

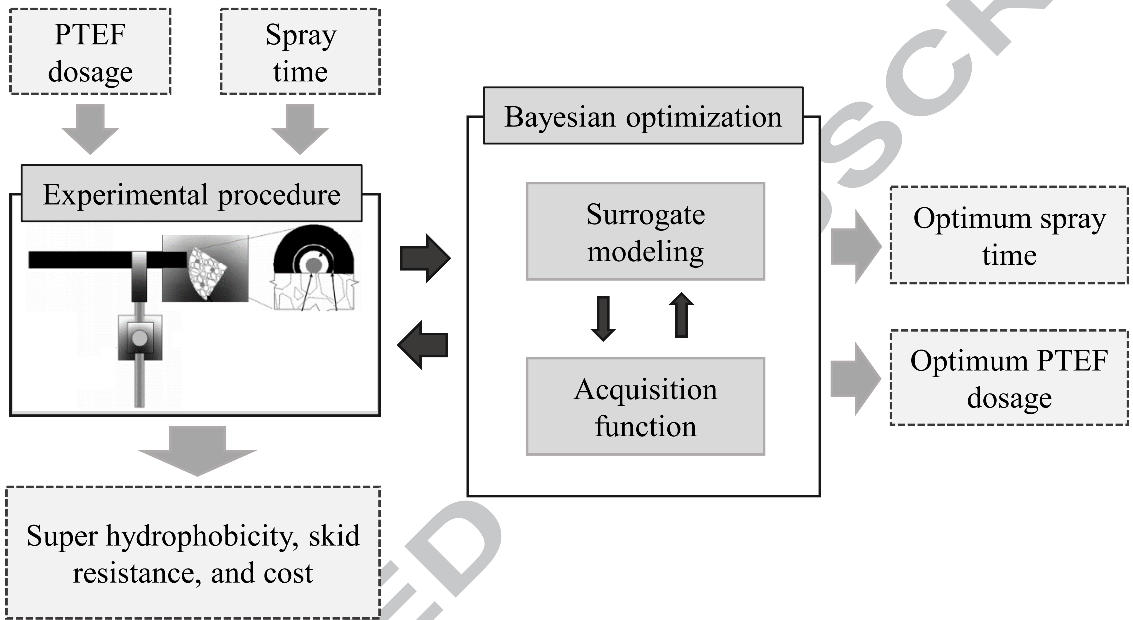


Figure 1 General procedure followed in this study.

Experimental procedure

The spray durations (3, 6, 9, and 12 s) and PTFE dosage rates (10%, 20%, 30%, and 40% which were all based on the weight of the two-part epoxy) were considered to be the initial set of input variables. To this end, twenty disk-shaped asphalt concrete substrates were made (see Figure 2b). Figure 2a presents the aggregate gradation used for fabrication of asphalt concrete substrates, and it is worth noting that aggregates act as skeleton is asphalt concrete. The asphalt binder, as its name indicates, is the component holding the aggregate system together. The physical properties of asphalt binder and aggregates are listed in Table 1.

Table 1 The physical properties of asphalt binder and aggregates

Aggregate	Specific gravity (g/cm3)	Absorption (%)
Limestone	2.76	1.44
Asphalt binder	Specific gravity (g/cm3)	Penetration value (0.1mm)
PG58-28	1.028	75

Note: PG = Performance Grade

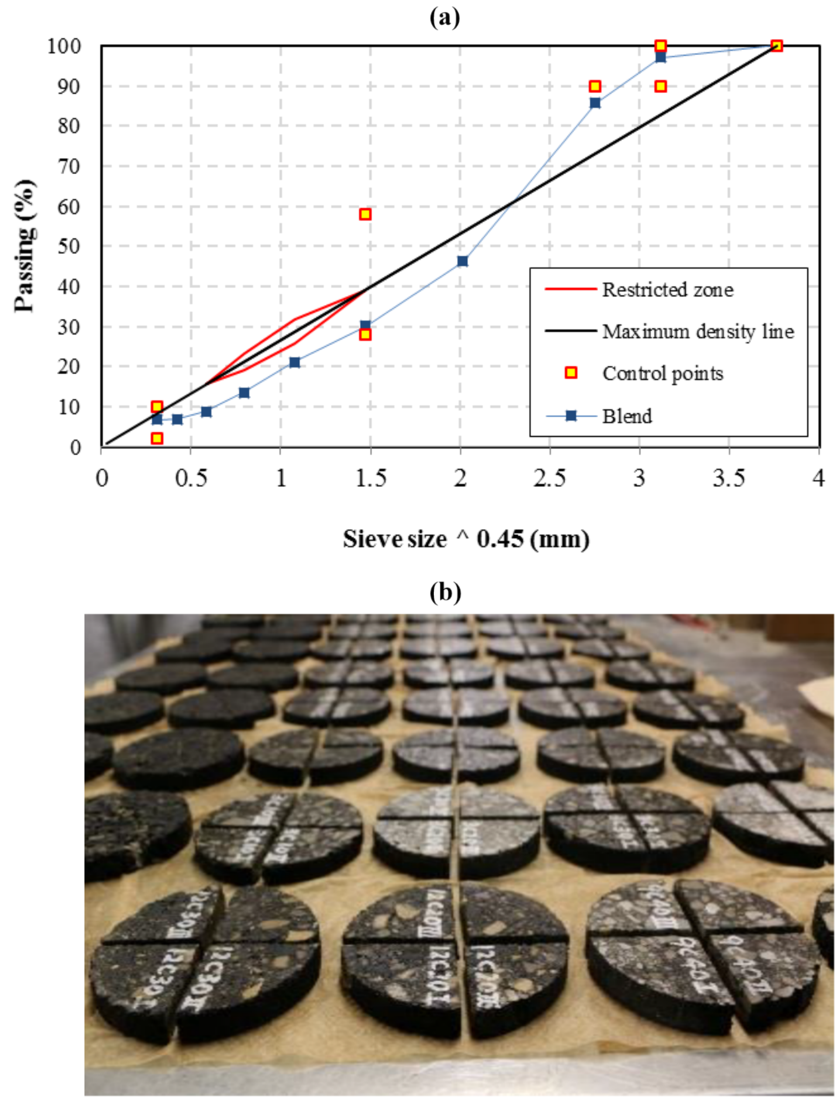


Figure 2 Substrate preparation: (a) selected aggregate gradation, (b) asphalt concrete substrates.

To obtain replicates, each disk was divided into four symmetrical quarters, i.e., 64 (16 × 4) replicates were coated using the LBL technique (Feng and Jiang 2006). In addition, 16 (16 × 1) samples were used for control. The reason behind selection of PTFE can be justified because of its promising water-repellency and tribological behavior that has been thoroughly explained in another study we conducted on tribological behavior and wettability of spray-coated superhydrophobic coatings (Young et al. 2017). According to the same study (Young et al. 2017), PTFE resulted in a satisfactory durability against grinding action which was performed by rubbing a 240-grit silicon carbide paper against the coated aluminum surfaces. In other words, there was not a considerable drop in water-repellency after reducing the coatings' thicknesses by approximately 30% (Young et al. 2017). In addition to the mentioned test, a ball-on-flat micro tribometer was used for performing reciprocal wear tests on the coatings by applying a contact pressure of 24 MPa. The results revealed that PTFE coating had a promising wear resistance (Young et al. 2017). The promising wear resistance of PTFE was in accordance with Bayer's

2017 comprehensive review paper (Bayer 2017), who conducted a thorough investigation on PTFE's wear resistance. According to Bayer, PTFE sustains superhydrophobicity until the PTFE material is completely worn away when rubbed with a fine grit sand paper. Also, it is worth noting that the durability of LBL coatings can be characterized using another novel approaches such as tape peel method (Bayer et al. 2016); however, such test method should be tailored for conditions that the coatings are exposed to heavy fast moving loads such as the ones applied by the wheels of vehicles/aircrafts. The asphalt concrete samples were composed of a dense-graded limestone aggregate blend and an optimal amount of unmodified PG 58-28 asphalt binder. PG 58-28 asphalt binder is a regular asphalt binder type suitable for southern Iowa's climatic conditions. The mix design used for fabricating the asphalt concrete samples was in conformance with the FAA advisory circular. The aggregate used was limestone with specific gravity and absorption of 2.76 g/cm^3 and 1.44%, respectively.

Because of the nonplanar surface of asphalt concrete, an LBL coating technique was used (Zhang et al. 2008) which is a top-down coating technique. In this method, at first, a binding layer is sprayed on the substrate and then the water-repellent material is spray deposited. In this study, the binding layer was EP 1224 epoxy that was purchased from ResinLab. This epoxy is commercially available in two parts: part A which is a polymer based resin and part B which is a curing agent. Part A was mixed with part B at the volume ratio of 2:1, and in order to decrease the viscosity, the epoxy was mixed with xylene and deposited for 3 s on the top surface of each asphalt concrete sample. The two-part epoxy was utilized to bind the PTFE to the asphalt concrete substrate. At that point, the PTFE (with the commercial name of Zynol MP 1300 PTFE and donated by DuPont with an average particle size of 12 microns) was dispersed in acetone and was sprayed over the epoxy resin in an amount based on the weight percentage of the epoxy resin. Figure 3 shows LBL deposit method used in this study.

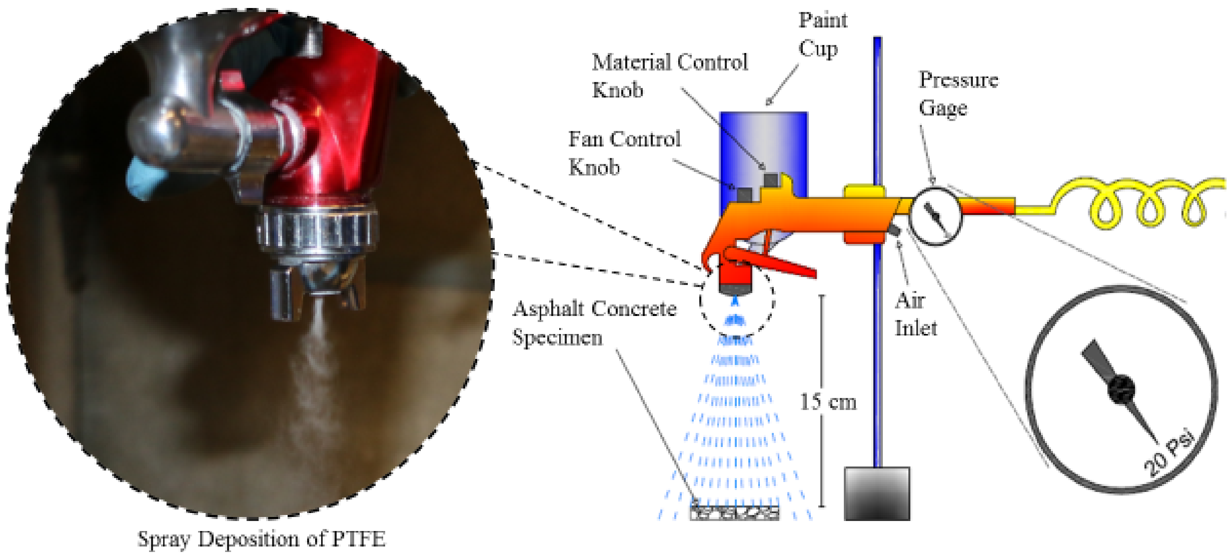


Figure 3 Experimental set-up.

Each coated specimen's superhydrophobicity was characterized by measuring the water contact angle by placing 4-μL water droplets with a micropipette on three spots distributed over the surface of each coated sample. The skid resistance of each sample was measured using a microtribometer-based CF measurement technique more completely described in a previous study (Arabzadeh et al. 2016c).

At the end, PTFE powder was characterized by scanning electron microscope (SEM) to investigate the particle size, microstructure, and the surface morphology of the particles. The specimens coated with PTFE-epoxy coating at different spray durations were also studied by SEM and energy dispersive X-ray spectroscopy (SEM/EDX). These specimens were coated with gold as a part of the SEM/EDX sample preparation procedure.

Bayesian Optimization (BO)

BO is a popular and computationally effective technique for solving optimization problems that involve assessment of costly, black-box objective (or response) functions. An underlying surrogate model is built over an objective function using kriging (or GP regression). In subsequent iterations, the surrogate model is sequentially updated using Bayes' rule (Mockus 1994; Nannapaneni et al. 2016; Zhao et al. 2011). Figure 4 shows the flowchart of a generic BO algorithm. The key step of the process involves finding a next sample point based on an acquisition function. Numerous acquisition functions have been developed to attain suitable trade-off between exploration and exploitation in an optimization problem. When appropriately applied, this trade-off balances the inspection of the areas in the design space with high uncertainty of model prediction (exploration) with focus on the regions that have seemed to approach the best optimum solution (exploitation) (Jones et al. 1998). In Figure , the level of convergence is defined as the amount of change in the predicted optimum point in two consecutive iterations (e_t). If e_t becomes less than a pre-defined threshold, e_0, then adding more points hardly changes our belief about the optimum solution and thus the predicted optimum point is deemed to converge to the optimum solution.

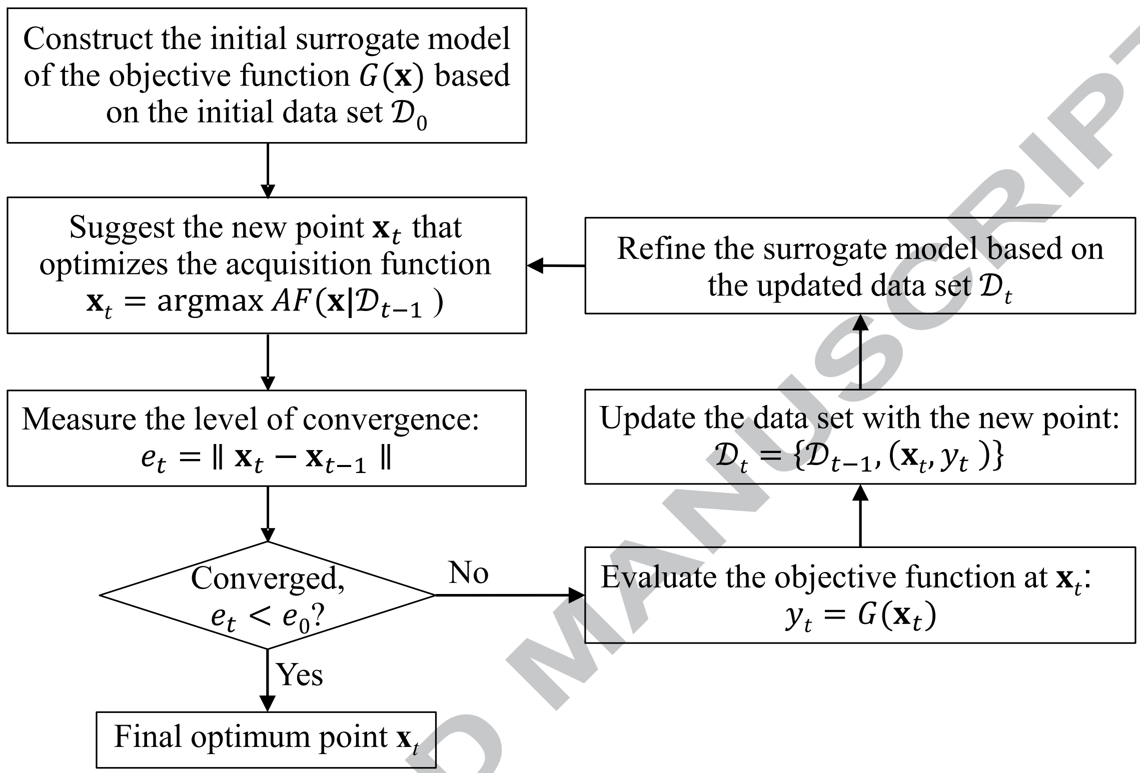


Figure 4 General algorithm of BO.

Multivariate Gaussian process

To model multiple (m) correlated outputs, a vector $\mathbf{Y}$ with $n_{\mathcal{D}}m$ elements is first built with the sample data set $\mathcal{D}$. The i^{th} set ($1 \le i \le n_{\mathcal{D}}$) of m elements of $\mathbf{Y}$ gathers the m outputs at the i^{th} input (or sample) point. For example, the first m elements of $\mathbf{Y}$ collect the m responses at the first input point, the second m elements gather the m outputs at the second input point, and the last m elements collect the m outputs at the $n_{\mathcal{D}}^{\text{th}}$ input point. Then, the vector $\mathbf{Y}$ follows a multivariate normal distribution whose density function takes the form:

$$f(\mathbf{Y}) = \frac{1}{(2\pi)^{n_{\mathcal{D}}m/2}\left(|\mathbf{\Sigma}_{\mathbf{Y}}|\right)^{1/2}} \exp\left[-\frac{1}{2}(\mathbf{Y}-\boldsymbol{\mu})^{\mathrm{T}}\mathbf{\Sigma}_{\mathbf{Y}}^{-1}(\mathbf{Y}-\boldsymbol{\mu})\right] \tag{2}$$

where $\boldsymbol{\mu}$ and $\mathbf{\Sigma}_{\mathbf{Y}}$ denote the mean vector and the covariance matrix of $\mathbf{Y}$, respectively. To derive the covariance matrix $\mathbf{\Sigma}_{\mathbf{Y}}$, we first define the correlation between the elements of the output vector $\mathbf{Y}$ (Mockus 1994). Here, we adopt the none-separable dependence model given in (Jones et al. 1998) that generates an m-variate Gaussian vector with mean vector $\boldsymbol{\mu}$ and covariance matrix $\mathbf{\Sigma}_{\mathbf{Y}}$ from a vector $\mathbf{Z}$ with m normally independently and identically distributed standard variables. The correlation between the responses at two candidate points $\mathbf{x}_i$ and $\mathbf{x}_{i'}$ is given by

$$Cov(\mathbf{Y}(\mathbf{x}_i), \mathbf{Y}(\mathbf{x}_{i'})) = \mathbf{A}\mathrm{diag}\left[\mathbf{R}(\mathbf{x}_i - \mathbf{x}_{i'};\boldsymbol{\theta}^{(1)}), \ldots, R(\mathbf{x}_i - \mathbf{x}_{i'};\boldsymbol{\theta}^{(m)})\right]\mathbf{A}^{\mathrm{T}} \tag{3}$$

where $\mathbf{R}$ is the Gaussian correlation function, a distance function expressed in the input dimensions, and $\boldsymbol{\theta}^i$ is a $d \times 1$ vector of hyperparameters that measures the importance of the inputs with respect to the i^{th} response and should be optimized before predicting the response at a set of candidate points $\mathbf{X}_c$. To this end, we apply the Cholesky transformation and estimate the hyperparameters by the restricted maximum likelihood estimation (Jones et al. 1998). Finally, the mean value of prediction and the mean squared prediction error (MSPE) are determined by

$$\hat{\mathbf{y}}(\mathbf{X}_c) = \hat{\boldsymbol{\mu}} + \boldsymbol{\Sigma}_{c,n_{\mathcal{D}}m}\boldsymbol{\Sigma}_{\mathbf{Y}}^{-1}(\mathbf{Y} - \mathbf{F}\hat{\boldsymbol{\mu}}) \tag{4}$$

$$\boldsymbol{\sigma} = \mathbf{MSPE}[\hat{\mathbf{y}}(\mathbf{X}_c)] = \hat{\boldsymbol{\Sigma}}_c - \hat{\boldsymbol{\Sigma}}_{c,n_{\mathcal{D}}m}\hat{\boldsymbol{\Sigma}}_{\mathbf{Y}}^{-1}\hat{\boldsymbol{\Sigma}}_{c,n_{\mathcal{D}}m}^{\mathrm{T}} + \mathbf{U}(\mathbf{F}^{\mathrm{T}}\hat{\boldsymbol{\Sigma}}_{\mathbf{Y}}^{-1}\mathbf{F})\mathbf{U}^{\mathrm{T}} \tag{5}$$

Here, $\boldsymbol{F} = \mathbf{1}_{n_{\mathcal{D}}} \otimes \mathbf{I}_m$ where $\mathbf{1}_{n_{\mathcal{D}}}$ denotes a $n_{\mathcal{D}} \times 1$ vector of ones, and $\mathbf{I}_m$ is an $m \times 1$ matrix of ones. $\boldsymbol{\Sigma}_c$ is the covariance matrix between the set of candidate points $\mathbf{X}_c$, and $\mathbf{U} = \mathbf{I}_m - \boldsymbol{\Sigma}_{c,n_{\mathcal{D}}m}$ $\boldsymbol{\Sigma}_{\mathbf{Y}}{}^{-1}\mathbf{F}$. For each candidate point, $\boldsymbol{\sigma}$ is a $m \times m$ covariance matrix that quantifies the correlation among the outputs (off-diagonal elements of $\boldsymbol{\sigma}$) and also the uncertainty in predction.

Acquisition Function

While researchers have previously developed a number of acquisition functions for problems involving single responses, no suitable acquisition function seems to exist for problems with multiple responses. With respect to the multivariate Gaussian outputs, this study develops a new acquisition function based on new definitions of the exploration and exploitation criteria. For the exploration criterion, a cross validation technique is adopted to quantify the nonlinearity of the objective function and prediction uncertainty over the design (or input) space, and for the exploitation criterion, the L2 norm of the mean prediction by multivariate GP is considered to measure the magnitude of the objective functions over the design space.

Conventionally, the leave-one-out cross validation (LOOCV) technique approximates the precision of a surrogate without requiring new testing points (Rana et al. 2017). In this study, LOOCV is instead used for exploring nonlinear and highly uncertain areas in the sequential sampling. The LOOCV technique basically removes one sample point in each assessment, and predicts the response at each candidate point using multivariate GP with the rest of data points. The prediction follows the joint Gaussian distribution, denoted by $\hat{\boldsymbol{\tau}}_{-i}(\mathbf{y}) \sim \boldsymbol{N}(\boldsymbol{\mu}_{-i}, \boldsymbol{\sigma}_{-i})$, where i is the index of the sample point removed from the data set. Then, the difference between the surrogate model built by removing the i^{th} sample point and the full surrogate model built with all sample points is quantified by a probabilistic divergence measure, namely the Kullback–Leibler divergence (Jones et al. 1998), that takes the following form

$$KL_{-i} = D_{KL}(\hat{\tau}_{-i}(\mathbf{y}) \,\|\, \hat{\tau}(\mathbf{y})) = \int \hat{\tau}_{-i}(\mathbf{y}) \log \frac{\hat{\tau}_{-i}(\mathbf{y})}{\hat{\tau}(\mathbf{y})} d\mathbf{y} \tag{6}$$

where $\hat{\boldsymbol{\tau}}(\mathbf{y}) \sim \boldsymbol{N}(\boldsymbol{\mu}, \boldsymbol{\sigma})$ denotes the probability density of the full surrogate model. For a multivariate Gaussian distribution, the above integral can be analytically solved as

$$KL_{-i} = \frac{1}{2}\left[\log \frac{|\boldsymbol{\sigma}_{-i}|}{|\boldsymbol{\sigma}|} - d + \mathrm{Tr}(\boldsymbol{\sigma}_{-i}^{-1}\boldsymbol{\sigma}) + (\boldsymbol{\mu}_{-i} - \boldsymbol{\mu})^{\mathrm{T}}\boldsymbol{\sigma}_{-i}^{-1}(\boldsymbol{\mu}_{-i} - \boldsymbol{\mu})\right] \tag{7}$$

The *KL* term on the left side of Eq. (6) quantifies, based on the i^{th} sample point, the nonlinearity of the objective function and the prediction uncertainty of the function over the design space. In this study, the total sum of squares of this term over all available sample points is considered as the exploration criterion. A proper acquisition function in BO should guide the search for optima towards regions with high nonlinearity and prediction uncertainty (exploration), and equally importantly, towards those with high values of prediction (exploitation). To this end, the L2 nom of the mean values $\boldsymbol{\mu}$ of the multivariate prediction at each candidate point is defined as the exploitation criterion. Overall, the acquisition function is expressed as:

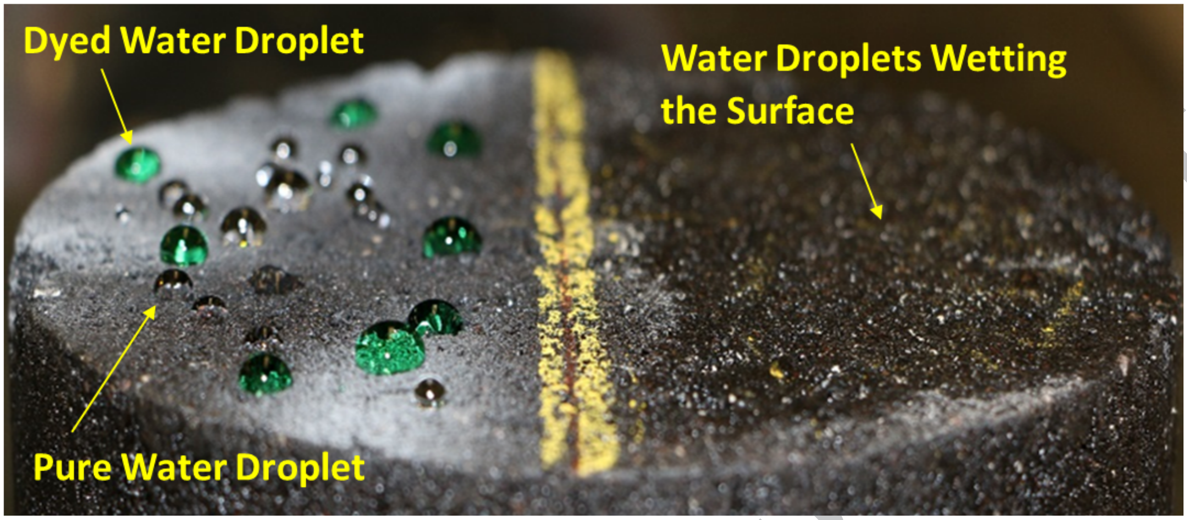
$$AF(\mathbf{x}_c) = \|\boldsymbol{\mu}\| \sqrt{\frac{1}{n_{\mathcal{D}}} \sum_{i=1}^{n_{\mathcal{D}}} \left[KL_{-i}(\mathbf{x}_c) \right]^2} \tag{8}$$

where $\|\cdot\|$ denotes the L2 norm and $n_{\mathcal{D}}$ is the number of sample points in $\mathcal{D}$. During the sequential sampling process in BO, the next single sample point $\mathbf{x}_t$ is suggested by maximizing the acquisition function, AF.

Results and Discussion

Figure 5(a) shows that the droplets form a round shape on the coated side (left side), indicating that they do not tend to wet the surface. The regular uncoated asphalt concrete shown on the right side of Figure 5(a), however, cannot form round shape droplets. Figure 5(b) also presents the water contact angles deposited on the curt surface of superhydrophobic asphalt concrete for the sake of water contact angle measurements. The contact angle of round shape droplets are equal to or larger than 150°, indicating that the surface is superhydrophobic.

(a)



(b)

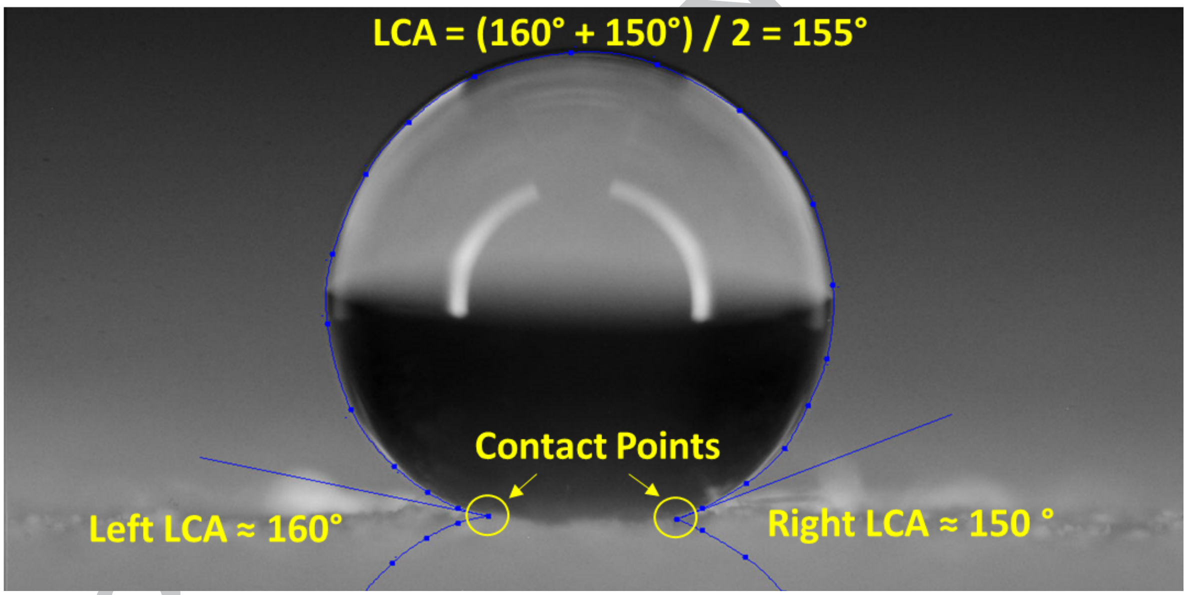


Figure 5 The behavior of water droplets on treated and untreated surfaces: (a) ≈ 60-μL water droplet on coated (left) and not coated (right) surfaces of asphalt concrete, (b) 4-μL water droplet deposited on a cut smooth superhydrophobic asphalt concrete surface.

In the next section, at the initial design of experiments, measured contact angle and CF will be analyzed, then the optimization algorithm will be applied on the initial test results to find the optimum and most cost-effective dosage rate and spray time.

Initial results of experiments

Water contact angle measurements as defined in the experimental procedure section were performed for each asphalt concrete sample. Table 2 gives the averaged values and standard deviations of water angle measurements for each combination of spraying time and PTFE dosage.

Table 2 Measured Contact Angles in Degrees for the initial values of spray time and dosage

Spray time	PTFE Dosage (%)							
	10%		20%		30%		40%	
	Average	STD	Average	STD	Average	STD	Average	STD
3	123	7.6	154	3.1	156	2.1	149	3.1
6	157	3.1	156	4.8	157	2.6	152	1.9
9	163	4.4	152	6.4	162	2.4	155	2.2
12	158	5.5	154	4.9	159	4.2	164	1.8

Spray time is an important variable affecting superhydrophobicity, and an increase in spray time from 3 to 6 s increased contact angles so they became greater than 150°. At spray durations of 9 and 12 s, superhydrophobicity remained, even though the super water-repellency did not consistently increment for every one of the specimens after a duration of 6-s. In addition, expanding the amount of PTFE, up to a specific level, in the coating of asphalt concrete similarly increased ice and snow repellency. At a spray time of 12 s and PTFE of 40%, the biggest contact angle of 164° is occurred. As mentioned in the introduction section, for airfield snow removal it is sufficient to achieve a superhydrophobic coated surface with 150° contact angles. Above that level (150° contact angles), an added amount of PTFE is uneconomical and does not significantly add to superhydrophobicity.

The results of the ramp load test were shown in Table 3. As it is shown in the table a growth in the spray duration from 3 to 6-s significantly increases CF values. In addition, as mentioned in the methodology section, to compare the skid resistance of the coated asphalt samples with uncoated samples, the CF was measured over three paths on 16 uncoated control specimens; each specimen had one replicate for control purposes. While an uncoated control sample also has a less skid-resistant surface than that for a 6 s spray time with more than a 10% PTFE dosage, the 10% PTFE dosage under different spray durations resulted in fewer skid-resistant surfaces than for the uncoated control sample.

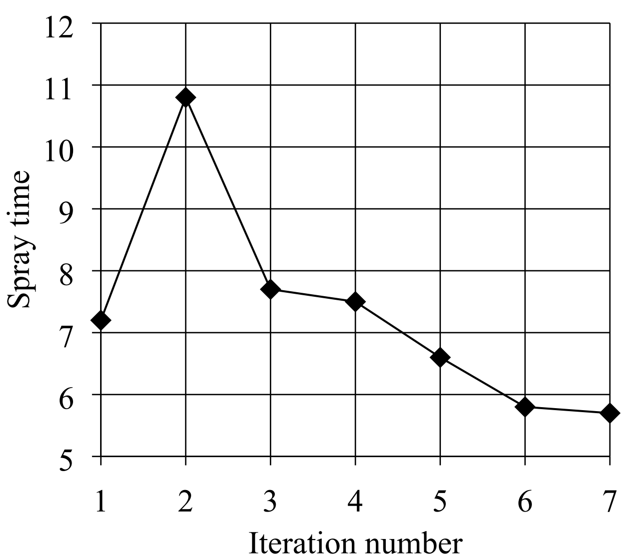
Table 3 Averaged CF values for the initial values of spray time and dosage

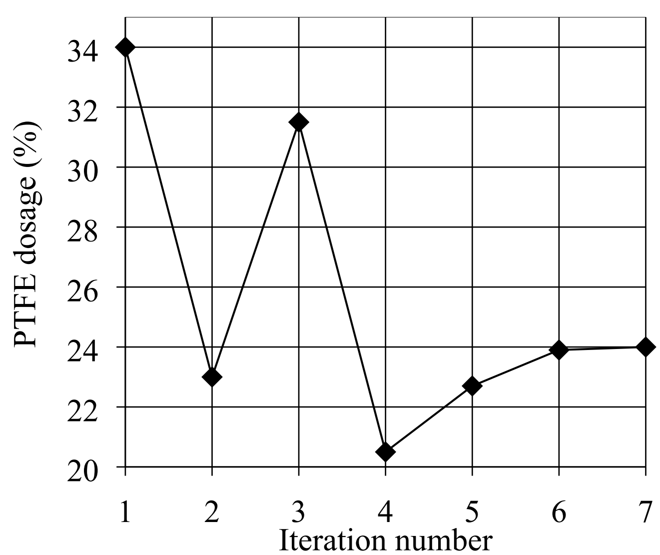
Spray time	PTFE Dosage (%)								Control	
	10%		20%		30%		40%			
	Average	STD	Average	STD	Average	STD	Average	STD	Average	STD
3	0.19	0.02	0.21	0.02	0.15	0.02	0.14	0.03	0.22	0.02
6	0.20	0.01	0.24	0.01	0.20	0.02	0.24	0.01		
9	0.14	0.01	0.23	0.02	0.23	0.01	0.26	0.01		
12	0.16	0.01	0.25	0.02	0.25	0.02	0.23	0.01		

Optimization results

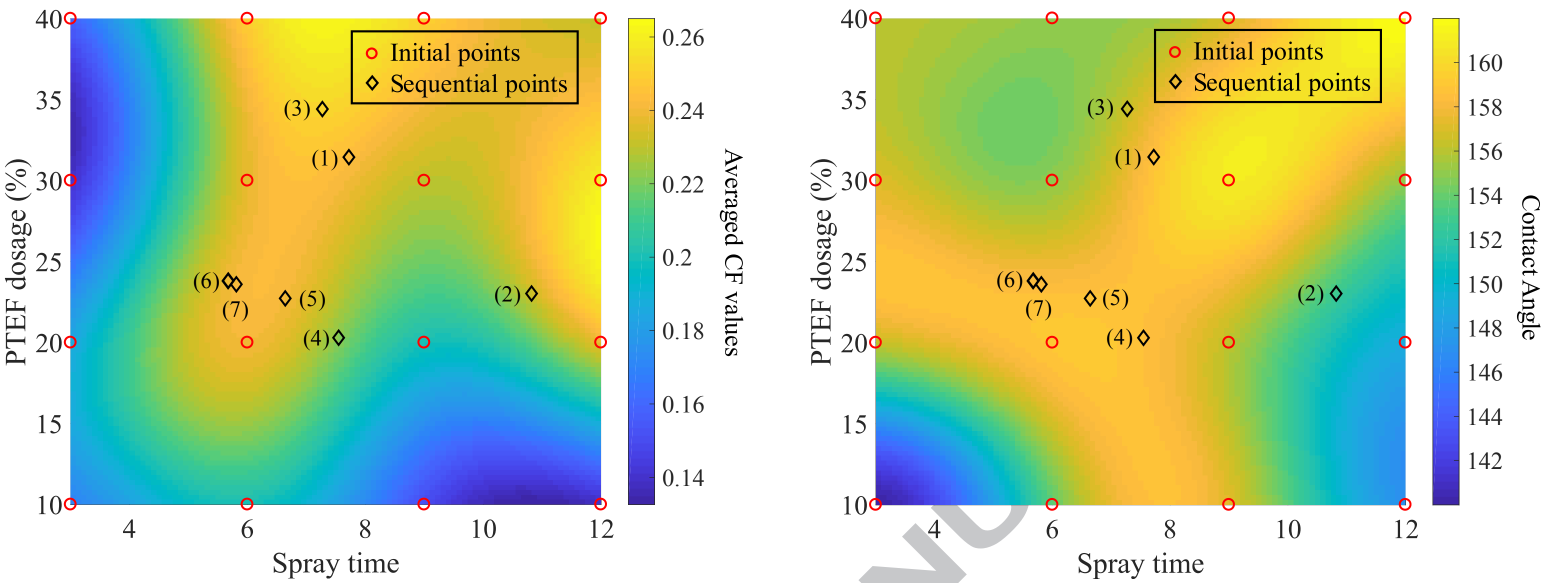
Based on the results from experiments at each initial sample point, an initial surrogate model was constructed over the entire range of the input variables. The surrogate model roughly estimates the behavior of the coefficient of friction and contact angle (superhydrophobicity), and using the acquisition function described in the methodology section, the accuracy of the surrogate model was improved sequentially by adding new sample points to the data set (Figure 6(a)). After achieving the proper model accuracy, the final sample point suggested by the acquisition function is considered to be the optimum design point. The hydrophobicity and skid resistance were maximally augmented using a dosage of approximately 23% PTFE for a spray time of 5.5-s. As mentioned before, one key part of the proposed optimization method is the utilization of multivariate GP instead of univariate GPs in surrogate construction. Multivariate GP is able to capture the correlations between the different objective functions by building a single joint surrogate. Due to the costly and time-consuming procedure of experiment, this study does not compare the performance between different optimization algorithms. However, the authors have recently demonstrated the outperformance of multivariate GP over univariate GPs in surrogate construction for system reliability analysis, when the response functions are highly correlated (Sadoughi et al. 2018).

Figure 6(b) shows the contours of hydrophobicity versus dosage-percentage and spray time. The coefficient of friction and contact angle varied over wide ranges, i.e., [0.14–0.26] and [142°–160 °], respectively. This figure shows the high interactive effects between the spray time and PTFE dosage on both outputs, indicating that, to maximize the CF and hydrophobicity, the effects of both dosage-percentage and spray time should be considered. At higher values of spray time and dosage, the CF and contact angle were maximized, implying that both hydrophobicity and skid resistance increase up to some specific points with increasing dosage percentage and spray time but, as mentioned in the methodology section, cost-effectiveness to provide affordable coating surfaces is important for agencies, so after considering the cost factor as an objective in the model the optimum point (point number 7) shown in Figure 6(b) was determined. This optimum point has similar CF and contact angle values to other points with higher values of spray time and dosage, but it represents lower material cost.





(a)



(b)

Figure 6 Interactive effects of spray time and dosage on CF and hydrophobicity; a) Sample points used at each iteration in the BO algorithm; b) Coefficient of friction (left) and contact angle (right side)

SEM imaging results

The SEM image of the particles are shown in Figure 7(a). It was found that PTFE powder comprised of micro-sized particles with surface roughness of approximately 200 nm. The majority of particles were in the size range of 5-10 microns. As seen in the figure, the PTFE particles were fully or partially embedded within the epoxy matrix that adhered them to the surfaces. The particles maintained their as-received morphology and surface structure in the coating layer.

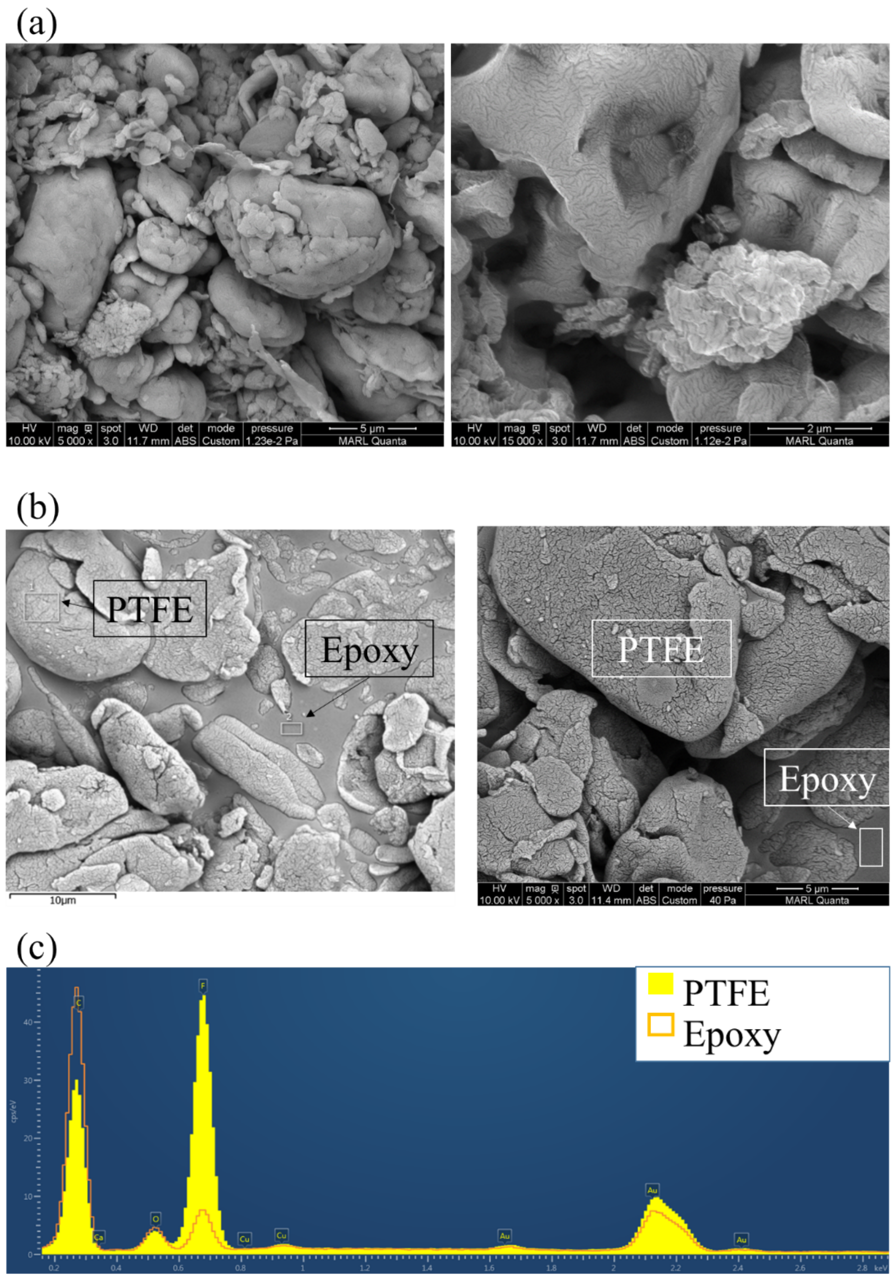


Figure 7 SEM imaging: (a) SEM images of as-received PTFE powder; (b) SEM images of PTFE-epoxy coating layer; and (c) X-ray spectra of PTFE and epoxy in the coating.

Low-magnification SEM images of coated specimens with different PTFE spray durations are shown in Figure 8. The images show that 6 seconds of spray duration resulted in the presence of a more uniform bed of PTFE on the surface of coating layer. Longer spray durations led to thicker layers of PTFE particles on the epoxy matrix, such that the amount of particles exceeded the capacity of the epoxy layer to hold them together. Therefore, some PTFE particles were detached from the surface and lost, resulting in more exposure of epoxy in the SEM images as seen in Figure 8.

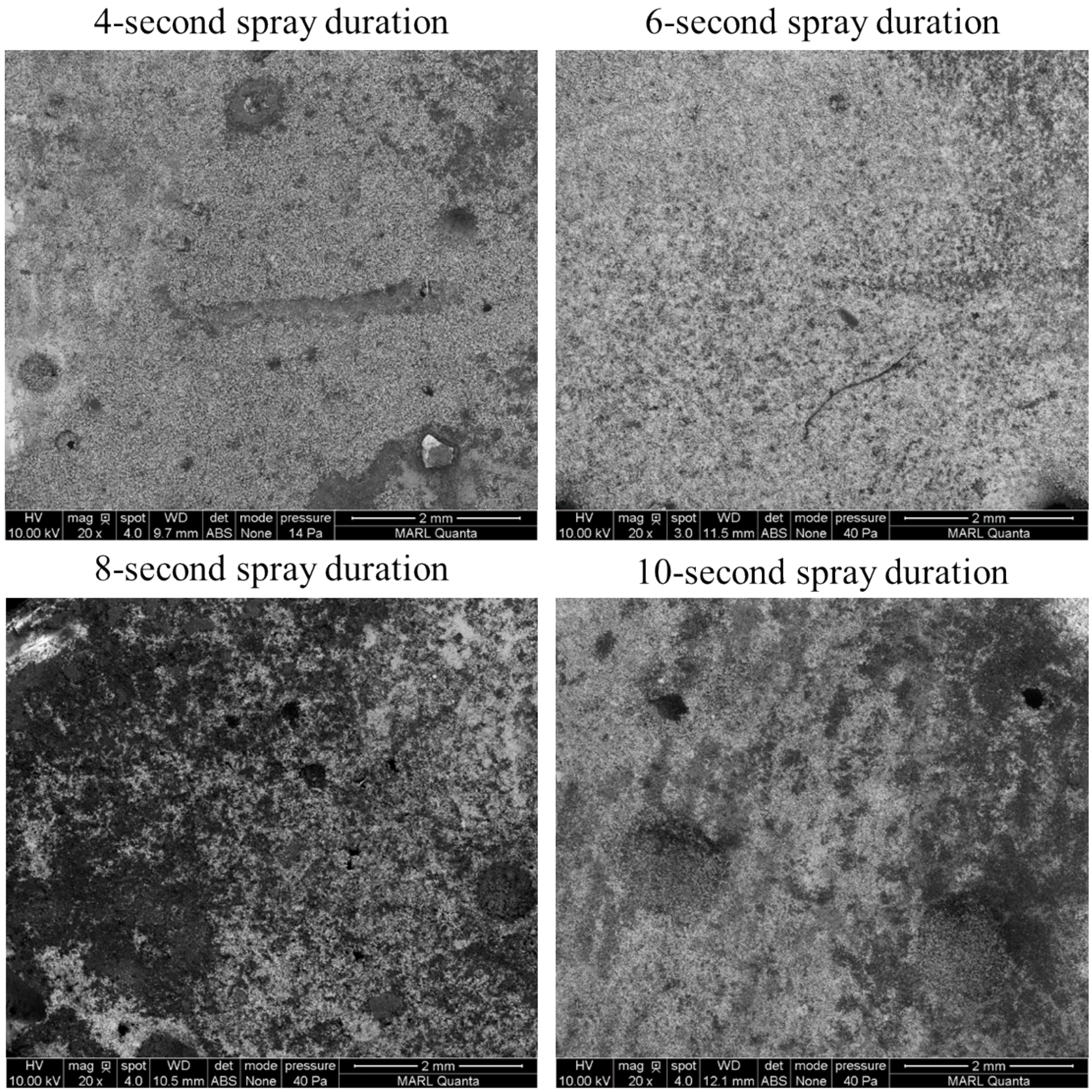


Figure 8 Low-magnification SEM/BSE images of PTFE-epoxy coatings with different PTFE spray durations.

Presence of weakly bonded PTFE particles on the surfaces of specimens coated by 8-seconds and 10-seconds PTFE spray duration is manifested in Figure 9 in the form of vast dark areas in their high-magnification SEM/BSE images. Some PTFE particles were detached from the surface after gold-coating exposing the underlying uncoated PTFE particles. These dark areas correspond to gold-lacking portions of the specimen caused by detachment of PTFE particles from the surface. The specimens coated at 4-second spray duration also exhibited a small amount of loose particles on the surface that were detached after sample preparation; this is probably because the particles were less strongly pushed within the epoxy matrix at shorter spray time. As seen in figures 8 and 9, the specimens coated at 6-second spray duration provided the most uniform distribution of PTFE and almost no particle loss from the surface either before or after gold coating. These observations set ground for concluding that applying 6-second spray duration, the amount of PTFE particles sprayed on the epoxy layer matched its particle accommodating capacity, while sufficient amount of PTFE particles were attained on the surface.

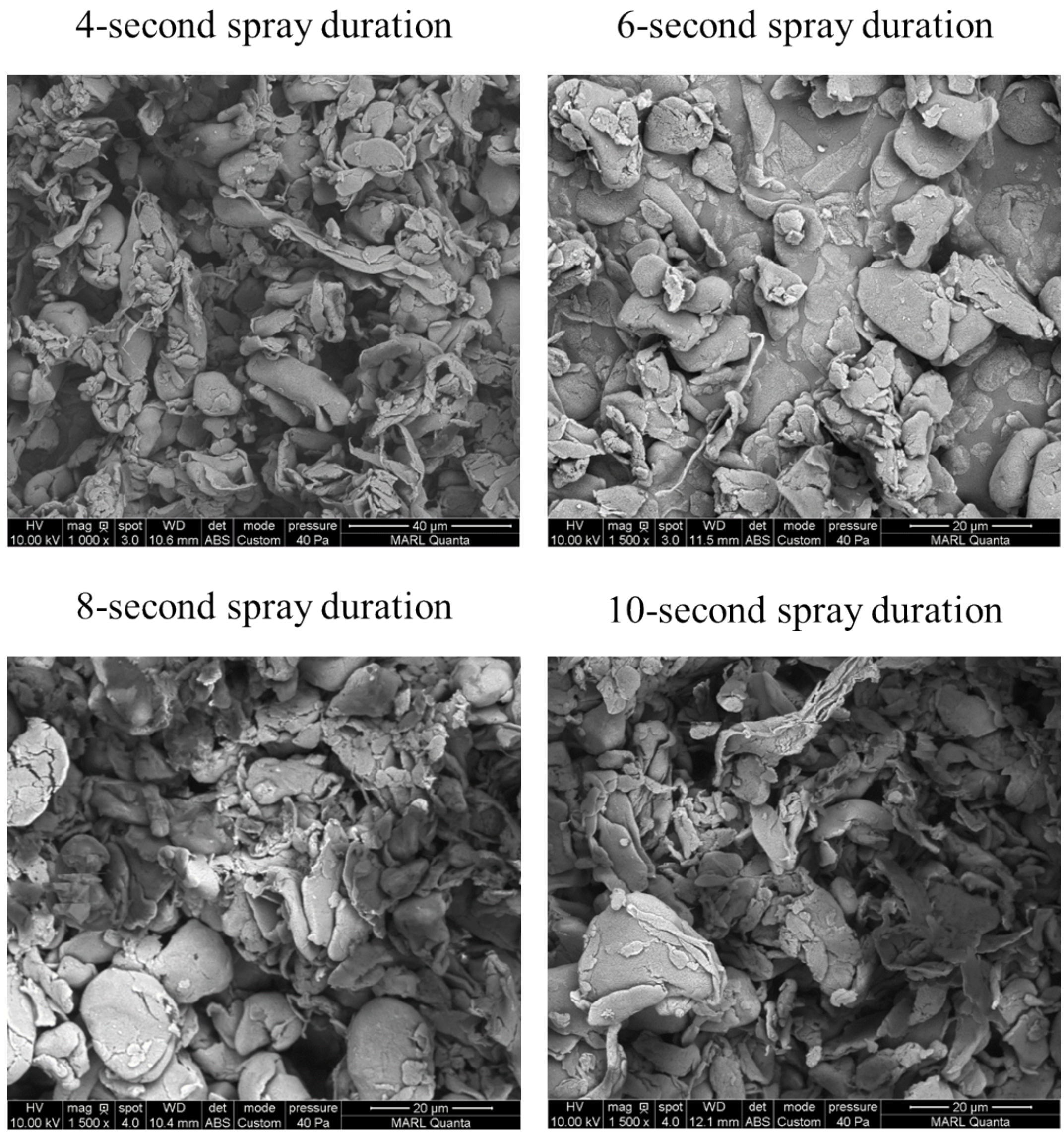


Figure 9 High magnification SEM/BSE images of PTFE-epoxy coatings with different PTFE spray durations.

Conclusion

This experimental and analytical study optimized hydrophobicity and skid resistance of coated asphalt concrete in a cost-effective manner. The LBL method and the microtribometer-based CF measuring method were used to coat asphalt concrete surfaces and to characterize the skid resistance of superhydrophobic coated substrates at the micro texture level. The optimum PTFE dosage and spray time that maximized superhydrophobicity and friction coefficient were determined using a multi-objective Bayesian optimization (BO) method. The highlights of the study can be summarized as follows:

- The developed multi-objective BO method properly modeled the interactive effects of spray time and dosage on the hydrophobicity and coefficient of friction (CF) using the minimum number of test points.
- The super water repellency and CF were not uniformly increased for all the specimens by increasing spray time and dosage.
- The hydrophobicity and skid resistance were maximally augmented by approximately 23% PTFE dosage, based on the weight of the two-part epoxy, at a spray time of 5.5 s.

In addition, the methodology followed in this study provides a computationally-effective technique for use in optimization problems that may require expensive, time-consuming, and labor-intensive experiments in which an outcome cannot be easily directly measured. This study did not consider the effect of durability on the results. For the future studies, it is recommended to investigate the effect of PTFE dosage and spray time on the durability of superhydrophobicity.

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Multi-objective Bayesian Optimization of Super hydrophobic Coatings on Asphalt Concrete Surfaces

Research Highlights

- Effects of spray time and dosage on the hydrophobicity and friction of asphalt were investigated.
- A layer-by-layer method was utilized for spray depositing polytetrafluoroethylene on an asphalt concrete.
- The optimum dosage and spay time were estimated by using a multi-objective Bayesian optimization method.
- An acquisition function that can tackle problems involving multiple objective functions was proposed.
- The optimum hydrophobicity and skid resistance were achieved with 23% PTFE dosage and at a spray time of 5.5s.